Mesoscopic Second-Harmonic Generation

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The low frequency second-harmonic transport response of multiprobe mesoscopic systems is analyzed using perturbation theory without explicit dephasing mechanisms. The intuitive conjecture that the second order transport current may be viewed as a linear conductance probing a distorted scattering potential is explicitly demonstrated in zero magnetic field **B**. For nonzero **B**, however, this simple picture breaks down, and $I(2\Omega)$ becomes a non-Fermi-surface quantity. A novel way of generating a second-harmonic transport current is predicted.

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The Landauer-Büttiker model has long been the paradigm describing linear dc transport in mesoscopic systems [1,2]. This framework separates a four-point resistance measurement on a length scale $L \lesssim L_\phi$ (the electron dephasing length) into quantum coherent propagation between leads and spatially separate dissipation occurring inside remote reservoirs. The linear conductance is then an algebraic combination of interlead transmission coefficients evaluated at the Fermi surface. The current through the sample depends only on the electrochemical potentials of the reservoirs, not on how the electrostatic potential varies between the leads. Recently there has been interest [3–9] in low frequency nonlinear mesoscopic transport. Guided by analogy with nonlinearities such as optical second-harmonic generation at surfaces, it may be anticipated that such measurements could probe phenomena inaccessible to the linear response. One aspect of nonlinear transport concerns low frequency second-harmonic generation, where a current $I(\Omega)$ at frequency Ω is imposed on a multiprobe mesoscopic system and the voltage response at 2Ω is measured. Although experiments of this type demonstrated the effect's quantum interference origins, they raised several unanswered questions: Is the quantity probed by second-harmonic generation restricted to properties at the Fermi surface? Can it be expressed in terms of equilibrium interlead transmission coefficients? Is it simply the linear conductance interrogating a distorted scattering potential, as assumed in some previous work [6,7]? How does the variation of the electrostatic potential enter and what is the role of Poisson's equation?

To resolve these issues it is constructive to look again towards nonlinear optics for a possible starting point [10]. These phenomena occur when the optical fields begin to alter the wave functions of the atoms being measured, and may be described by a perturbation method. An approach similar in spirit is taken here. Still retaining the assumption of spatially separate coherent propagation and dissipation in reservoirs, a microscopic linear response (Kubo) formalism [11] is extended to second order. This treatment is a natural extension of the first order calculation, which explicitly derived the Landauer-Büttiker picture. At the same time the spatial variation of the electrostatic potential is handled self-consistently. This differs from previous theoretical investigations of nonlinear transport. The surprise is that only for $\mathbf{B}=0$ is the second order ac response a Fermi-surface quantity that can be viewed in terms of distorted interlead transmission coefficients. This is due to a volume current generation mechanism derived below.

Figure 1 illustrates the model of Baranger and Stone adopted here [11]. A sample of volume V is connected to infinitely long disorder-free leads serving as reservoirs. The unperturbed Hamiltonian is of the single electron form: $\hat{H}_0=(1/2M)[(\hbar/i)\nabla-(e/c)\mathbf{A}]^2+U(\mathbf{r})$ with eigenfunctions $\psi_n(\mathbf{r})$ at continuous energy levels ε_n. In V, U corresponds to some disordered potential, while in the leads $\partial U/\partial x=0$, where $\hat{x}$ is outgoing along the lead. **A** describes the static magnetic field. For simplicity, explicit dephasing is not included. The perturbing Hamiltonian is taken as $\hat{H}[\phi]=\int d1\,\hat{\rho}(1)\phi(1,t)$, where $\hat{\rho}$ is the charge-density operator, ϕ is the *total* (screened) electrostatic potential, and the spatial variable $1\equiv\mathbf{r}_1$ ranges here over V and the leads. The leads are taken as perfect conductors so $\phi=\Phi_i$, a constant on lead i. Capacitor plates situated where $\psi(r)=0$ vary the electric field inside V in a manner consistent with the applied Φ_i.

The external potentials on the leads and plates, which are all grounded ($\phi=0$) in equilibrium, oscillate as $\cos\Omega t$ with a typical experimental frequency $\Omega\leq10^3$ Hz. Since Ω is less than typical wave-packet traversal rates across micron-sized samples and dephasing rates ($>10^8$ Hz), the $\Omega\rightarrow0$ limit is appropriate. The density matrix

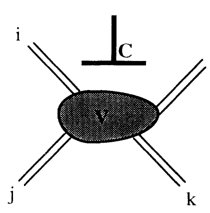

FIG. 1. A schematic of the model used to analyze the *second* order mesoscopic current response. The potential $U(r)$ is some arbitrary disordered potential inside the sample volume (V) and also confines the electrons to the ideal disorder-free semi-infinite leads (i,j,k,l). The electrostatic potential is assumed constant in each lead and the electrons experience $\mathbf{E}\neq0$ only in V. A nearby capacitor plate (C) is also shown.

is expanded in powers of $\hat{H}$ as $\hat{f}=\hat{f}_0+\hat{f}_1[\phi]+\hat{f}_2[\phi,\phi]+\cdots$, where $\hat{f}_0$ is the Fermi function describing global equilibrium. ϕ must obey the self-consistency requirement $\nabla^2\phi(r)=-4\pi\mathrm{Tr}\hat{f}\hat{\rho}(r)$. Using the expansion of $\hat{f}$ one can write the charge-density response in powers of $\hat{H}$ to generate $\phi=\phi_1+\phi_2+\cdots$, corresponding to self-consistent screening to first, second, and higher order in $\hat{H}$. This gives

$$\nabla^2\phi_1(r)=-4\pi\mathrm{Tr}\{\hat{f}_1[\phi_1]\hat{\rho}(r)\}\,, \tag{1}$$

$$\nabla^2\phi_2(r)=-4\pi\mathrm{Tr}\{\hat{f}_1[\phi_2]\hat{\rho}(r)+\hat{f}_2[\phi_1,\phi_1]\hat{\rho}(r)\}\,. \tag{2}$$

We are interested in the current response

$$\langle J_\alpha(0,t)\rangle=\mathrm{Tr}\{F_0+F_1+F_2+\cdots\}\,, \tag{3}$$

$$F_0=\hat{f}_0\hat{j}_\alpha(0)\,,$$

$$F_1=\hat{f}_1[\phi_1]\hat{j}_\alpha(0)\,,$$

$$F_2=(\hat{f}_2[\phi_1,\phi_1]+\hat{f}_1[\phi_2])\hat{j}_\alpha(0)\equiv A+B,$$

and particularly in the second-harmonic transport current up lead i, $I_i(2\Omega)=\int dS_{0i\alpha}\langle J_\alpha(0)\rangle$, the surface integral in $\mathbf{r}_0$ being over a cross section of lead i. F_0 describes equilibrium (dc) persistent currents, which do not contribute to transport. F_1 represents the linear response generated by the *first* order potential distorting wave functions to *first* order [11]. F_2 generates currents at 2Ω and dc. Within this interesting nonlinear term, A corresponds to currents produced by the *first* order potential distorting wave functions to *second* order, and B to currents arising from the *second* order potential acting to *first* order.

Linear response corresponds to solving the first order time evolution equation $i\hbar\,\partial\hat{f}_1/\partial t=[\hat{H}_0,\hat{f}_1]+[\hat{H}_1,\hat{f}_0]$, with $\hat{f}_1=\hat{f}_1[\phi_1]$ and $\hat{H}_1=\hat{H}[\phi_1]$, to compute $I_i(\Omega)$. To calculate $I_i^B(2\Omega)$ we replace ϕ_1 in $f_1[\phi_1]$ by ϕ_2. ϕ_2 and $I_i^A(2\Omega)$ both require $\hat{f}_2[\phi_1,\phi_1]$ from the second order equation of motion: $i\hbar\,\partial\hat{f}_2/\partial t=[\hat{H}_0,\hat{f}_2]+[\hat{H}_1,\hat{f}_1]$, with $\hat{f}_2=\hat{f}_2[\phi_1,\phi_1]$ and $\hat{f}_1=\hat{f}_1[\phi_1]$. $I_i^A(2\Omega)$ is computed as $\Omega\to 0$ to yield [12]

$$\langle J_\alpha^A(2\Omega,0)\rangle=\int_V d1\,d2\,E_\beta(1)E_\gamma(2)\sigma_{\alpha\beta\gamma}(0,1,2)+E_\alpha(0)\int_V d1\,E_\beta(1)Y_\beta(0,1)\,, \tag{4}$$

$$\sigma_{\alpha\beta\gamma}(0,1,2)=\kappa_\sigma\sum_{m,n,p}W_{np}(0)_\alpha W_{mn}(1)_\beta W_{pm}(2)_\gamma\left\{\frac{f_{mn}}{\varepsilon_{mn}}\left[\frac{P}{\varepsilon_{np}}-i\pi\delta(\varepsilon_{np})\right]\frac{\partial}{\partial\varepsilon_m}\left[\frac{P}{\varepsilon_{mn}}+i\pi\delta(\varepsilon_{mn})\right]\right.$$
$$\left.+\frac{f_{mp}}{\varepsilon_{mp}}\left[\frac{P}{\varepsilon_{np}}-i\pi\delta(\varepsilon_{np})\right]\frac{\partial}{\partial\varepsilon_m}\left[\frac{-P}{\varepsilon_{mp}}+i\pi\delta(\varepsilon_{mp})\right]\right\}, \tag{5}$$

$$Y_\beta(0,1)=\kappa_Y\sum_{m,n}W_{mn}(1)_\beta\psi_n^*(0)\psi_m(0)\frac{f_{mn}}{\varepsilon_{mn}}\left[\frac{P}{\varepsilon_{nm}}-i\pi\delta(\varepsilon_{nm})\right]. \tag{6}$$

Here m,n,p label complete eigenstates of $\hat{H}_0$, P denotes principal value, $\varepsilon_{mn}\equiv\varepsilon_m-\varepsilon_n$, $f_{mn}\equiv f_0(\varepsilon_m)-f_0(\varepsilon_n)$, $W_{mn}(1)_\beta\equiv(-2M/ie\hbar)\langle m|\hat{j}_\beta(1)|n\rangle$, and $\kappa_\sigma=(-i/16)(e\hbar/M)^3\hbar^2$, $\kappa_Y=(-i/4)(e\hbar)^3/M^2$. $E_\alpha(0)\equiv-\nabla_\alpha\phi_1(0)$ is the *first* order electric field in V, and the spatial integrals are over V only. Since $\mathbf{E}=0$ in the leads, integrating twice by parts gives

$$I_i^A(2\Omega)=I_i^{\mathrm{SS}}+I_i^{\mathrm{SV}}+I_i^{\mathrm{VV}}\,, \tag{7}$$

$$I_i^{\mathrm{SS}}=\sum_{j,k}\Phi_j\Phi_k\int\sigma_{\alpha\beta\gamma}(0,1,2)dS_{0i\alpha}dS_{1j\beta}dS_{2k\gamma}\,,$$

$$I_i^{\mathrm{SV}}=-\sum_k\Phi_k\int_V d1\,\phi_1(1)\int dS_{0i\alpha}dS_{2k\gamma}[\sigma_{\alpha\beta\gamma,\beta}(0,1,2)+\sigma_{\alpha\gamma\beta,\beta}(0,2,1)]\,,$$

$$I_i^{\mathrm{VV}}=\int_V d1\,d2\,\phi_1(1)\phi_1(2)\int dS_{0i\alpha}\sigma_{\alpha\beta\gamma,\beta\gamma}(0,1,2)\,.$$

Here the subscript β denotes $\partial/\partial r_{1\beta}$. The three terms on the right-hand side of (7) are designated as the surface-surface (SS), surface-volume (SV), and volume-volume (VV) contributions according to how the two potential factors enter: as a surface integral at a lead or as a volume integral over V. As a result of current conservation, the surface integrals may be evaluated anywhere along the leads.

Under constant *current* bias conditions [3,4] where $I_s(\Omega)=-I_d(\Omega)=I$, $I_k(\Omega)=0$ for leads $k\neq s,d$, and $I_k(2\Omega)=0$ for all k, the imposed current at Ω generates the reservoir potentials $\Phi_k(\Omega)$ through the linear conductance G_L. The reservoir and capacitor potentials provide boundary conditions for Eq. (1) with solution $\phi_1(r,\Omega)$. Because $I_i^B(2\Omega)$ is the first order current response to ϕ_2, $I_i^B(2\Omega)=\sum_j(G_L)_{ij}\Phi_{2j}(2\Omega)$, where $\Phi_{2j}(2\Omega)$ is $\phi_2(r,2\Omega)$ for r on lead j. Since $I_i^A(2\Omega)+I_i^B(2\Omega)=0$, we have $\Phi_i(2\Omega)=-\sum_j(G_L^{-1})_{ij}I_j^A(2\Omega)$, where I_j^A is given by (7) with $\Phi_k=\Phi_k(\Omega)$, and there is no need to solve (2) [13].

To relate $I_i(2\Omega)$ to transmission amplitudes we introduce the Green's functions

$$G_\varepsilon^\pm(0,1)=\lim_{\eta\to 0+}\sum_n\psi_n(0)\psi_n^*(1)/(\varepsilon-\varepsilon_n\pm i\eta)\,,$$

$$\Delta G_\varepsilon(0,1)=G_\varepsilon^+(0,1)-G_\varepsilon^-(0,1)\,,$$

$$\Sigma G_\varepsilon(0,1)=G_\varepsilon^+(0,1)+G_\varepsilon^-(0,1)\,.$$

For example, as $x_0,x_1\to\infty$ in leads $j\neq k$, $G(0,1)$ can be related to the scattering amplitude from lead k to lead j [11]. The result for the VV current is

$$I_i^{\mathrm{VV}}(2\Omega)=|\kappa_\sigma|(1/2)^2(1/2\pi)(2M/\hbar^2)^2\int_V d1\,d2\,\phi_1(1)\phi_1(2)\int dS_{0i\alpha}\int d\varepsilon f_0(\varepsilon)\times\mathrm{Re}\{\Delta G_\varepsilon(2,1)\Sigma G_\varepsilon(1,0)\overleftrightarrow{D}(0)_\alpha\Sigma G_\varepsilon(0,2)\}\,. \tag{8}$$

Here

$$g(0)\overleftrightarrow{D}(0)_\alpha h(0) \equiv g(0)\{[\partial/\partial r_{0\alpha} - (ie/\hbar c)A_\alpha(0)]h(0)\} - \{[\partial/\partial r_{0\alpha} + (ie/\hbar c)A_\alpha(0)]g(0)\}h(0).$$

The SS and SV currents yield much lengthier expressions that also contain energy derivatives of $f_0(\varepsilon)$ besides terms like VV without such derivatives. Equation (1) may be reduced to a linear inhomogeneous integral equation for ϕ_1 [12]. Fortunately it is not necessary to delve into it to grasp the essential physics.

Linear response [11] yields $I_i(\Omega) = \sum_j (G_L)_{ij}\Phi_j(\Omega)$, where G_L involves interlead transmission amplitudes near the Fermi surface. For $I(2\Omega)$, $I_i^{\mathrm{SV}} + I_i^{\mathrm{VV}} \neq 0$ even for $\mathbf{B}=0$, so $I_i(2\Omega) \neq \sum_{j,k} G_{ijk}\Phi_j\Phi_k$. In contrast to the linear response, the second order current also depends on how the potential drops through V. Because I^{SV} and I^{VV} involve propagation to points inside V (see Fig. 2), the response cannot be written in terms of *equilibrium* interlead transmission amplitudes or their energy derivatives. The insensitivity of the linear conductance to how $\phi(r)$ varies inside V arises from the severe constraints imposed by current conservation and time-reversal symmetry [11]. These requirements are not as restrictive regarding $I(2\Omega)$. Unlike the linear response, $I(2\Omega)$ is not a simple commutator and so does not obey the Onsager-Büttiker symmetries [2,14].

For $\mathbf{B}\neq 0$, $I(2\Omega)$ contains off-Fermi-surface (OFS) contributions arising from terms without an energy derivative of $f_0(\varepsilon)$. A simple physical argument explains why $I(\Omega)$ is a Fermi-surface quantity while $I(2\Omega)$ is not. Consider linear transport as an electron incident in state $|i\rangle$ at energy ε_i interacting *once* with $\phi(r)$ and emerging in $|f\rangle$ at energy ε_f. Since total energy is conserved and to maintain phase coherence, one must have $\varepsilon_f = \varepsilon_i = \mu_s$ at $T=0$ where μ_s is the source reservoir's electrochemical potential. $|f\rangle$ later relaxes to the drain electrochemical potential within the drain lead. When two scatterings off $\phi(r)$ occur, generating $I(2\Omega)$, $|i\rangle$ first scatters to an intermediate state $|v\rangle$ which then scatters to a final state $|f\rangle$ with $\varepsilon_f = \varepsilon_i$. $|v\rangle$, however, need not be at this energy. As $\Omega \to 0$ energy conservation may be violated at each vertex so long as overall energy conservation is maintained.

The perturbation approach also answers whether the second order response can be viewed as a linear conductance probing scattering off a distorted potential. This interpretation separates the two actions of ϕ into inequivalent roles: One distorts the scattering potential and the other probes it through modified transmission amplitudes. For this to be true, one must demonstrate that, as $\Omega \to 0$,

$$I_i(2\Omega) \propto \sum_{j\neq i} \Phi_j \int dS_{0i\alpha} dS_{1j\beta} \int d\varepsilon f'(\varepsilon)\delta[G_\varepsilon^+(0,1)\overleftrightarrow{D}(0)_\alpha^* \overleftrightarrow{D}(1)_\beta G_\varepsilon^-(1,0)] - \tfrac{1}{2}\Phi_i \int dS_{0i\alpha} dS_{1i\beta} \int d\varepsilon f'(\varepsilon)\delta[\Delta G_\varepsilon(0,1)\overleftrightarrow{D}(0)_\alpha^* \overleftrightarrow{D}(1)_\beta \Delta G_\varepsilon(1,0)], \quad (9)$$

which is written in the form of a modified linear current [11] where δ represents the distorting action. For the second order case,

$$\delta[G^+(0,1)D(0)^*D(1)G^-(1,0)] = \delta G^+(0,1)D(0)^*D(1)G^-(1,0) + G^+(0,1)D(0)^*D(1)\delta G^-(1,0),$$

where the δG correspond to first order perturbed (nonequilibrium) propagators [15]. These δG can be written as $\int_V d2E_\gamma(2)\times$(products of G). Using $E(2)_\gamma = -\phi_{,\gamma}(2)$, and integrating by parts, we see that a second order response of the form (9) contains SS and SV terms but cannot generate a VV current. Time-reversal symmetry dictates that the VV term (8) is antisymmetric in the magnetic field $\mathbf{B}$, and $I_i(2\Omega)$ does take the form (9) for $\mathbf{B}=0$. For $\mathbf{B}\neq 0$, however, the second order current cannot be viewed as scattering off a modified potential. Most importantly, the VV term implies that theories of ac nonlinear mesoscopic transport expressed as modified transmission coefficients [6,7] are limited to zero magnetic field.

Figure 2 displays a physical description of these results. In linear response current conservation and time-reversal invariance require that $I_i(\Omega)$ depends only on the lead potentials. Figure 2(a) depicts how the linear currents can be interpreted as ϕ causing a charge buildup via electron-hole creation in lead j. These propagate across V, and a current flows up lead i. The SS, SV, and VV pieces of $I(2\Omega)$ involve two actions of ϕ [Figs. 2(b)-2(e)]. VV has ϕ acting twice in the *interior* of V. One of the two contributions to VV is shown in Fig. 2(d), where $\phi(1)$ creates an electron-hole pair, $\phi(2)$ subsequently scatters the electron, and then current is projected up lead i. Figure 2(e) shows the other part of VV which represents the current being generated first accompanied by pair creation, followed by the electron scattering off $\phi(2)$, and finally $\phi(1)$ absorbs the pair [time reverse of Fig. 2(d)]. In the absence of a magnetic flux threading V these two counterpropagating currents cancel, but for $\mathbf{B}\neq 0$ oppositely signed Aharonov-Bohm phases produce a nonzero sum. Such VV currents cannot be viewed as distorted linear currents since $I(\Omega)$ arises purely from charge pileup in the leads. The OFS contributions to the SS and SV terms also arise from similar processes that cancel for $\mathbf{B}=0$. Therefore the VV currents grow to the size of the other currents when a flux of order hc/e threads V.

The presence of the VV term suggests an intriguing experiment. Suppose all the leads connected to V are biased at ground potential while a nearby capacitor voltage oscillates at Ω. Then $I_i^A(2\Omega)=I_i^{VV}(2\Omega)\neq 0$ for nonzero fields, and $I_i^B(2\Omega)=\sum_j (G_L)_{ij}\Phi_{2j}(2\Omega)=0$. Consequently, a second-harmonic current flows in the leads generated only by the changing potential profile inside V without any analogous linear current. The order of magnitude of this "volume generated current" is roughly the same as the corresponding second-harmonic current generated by biasing the leads.

On the other hand, if such an experiment were to be performed *strictly at dc* ($\Omega=0$), then no transport currents would flow even for $\mathbf{B}\neq 0$ because then one simply has a new equilibrium Hamiltonian $\hat{H}_0$ with a modified potential U inside V. The above perturbation analysis, however, only computes the response to $\hat{H}$ through second order, ignoring higher order processes. All of these must be summed to obtain the strict dc response. When can these higher order terms be neglected? The perturbation approach is an expansion in the number of interactions an electron experiences with the oscillatory electrostatic potential while crossing V. The probability η of such "photon" absorption must be small. η is related to the cycle-averaged energy dissipation rate $\bar{P}$ by $\eta=(\bar{P}/\hbar\Omega)\hbar/E_c$, where $\hbar/E_c$ is the time for a wave packet to traverse V. For the capacitor-biased experiment sketched above, $\eta\sim\phi^2(2\Omega)G_L/2\Omega E_c$. $\eta\ll 1$ defines a region in the $\phi(\Omega)$-Ω plane where perturbation theory holds. This imposes the constraint

$$\phi(\Omega)\leq\phi_{\max}\sim(3.4\,\mu\mathrm{V})(\eta/0.01)^{1/4}[G_L^{-1}/(3\,\mathrm{k}\Omega)]^{3/4}[V_c/(21\,\mu\mathrm{V})]^{1/4}[f/(1\,\mathrm{kHz})]^{1/4},$$

where $E_c=eV_c$ and $\Omega=2\pi f$, for heterostructure samples [4]. This yields a current up the leads at 2Ω of order

$$(0.37\,\mathrm{pA})[\phi(\Omega)/\phi_{\max}]^2(\eta/0.01)^{1/2}[G_L^{-1}/(3\,\mathrm{k}\Omega)]^{-1/2}[V_c/(21\,\mu\mathrm{V})]^{1/2}[f/(1\,\mathrm{kHz})]^{1/2}.$$

The expansion fails when $\eta>1$; then the strong (multiphoton) interaction with ϕ "dresses" $\hat{H}_0$. Perturbation theory also requires only slightly distorted occupation numbers: $\zeta\equiv\langle n|f_1|n\rangle\ll\langle n|f_0|n\rangle\approx 1$. Computing $\bar{P}$ for the $T=0$ two-lead case yields $\zeta\sim e\phi(\Omega)/E_c$, in accordance with Altshuler, Khmelnitskii, and Larkin [5].

In conclusion, a perturbation analysis of mesoscopic second-harmonic current transport has revealed a new volume generation (VV) mechanism that has no analog in linear response. For nonzero $\mathbf{B}$, $I(2\Omega)$ contains off-Fermi-surface terms, and because of the double interior action of the electric potential, the commonly held view that the second order ac response can be pictured as a linear conductance probing a distorted interlead scattering potential is incomplete. Experiments are underway to explore these phenomena directly.

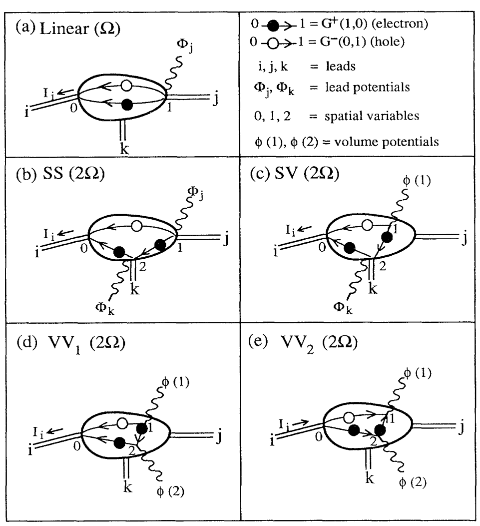

FIG. 2. Upper right panel defines symbols. (a) Schematic of linear current generation as charge buildup (electron-hole pair creation) in lead j, propagation across V, and final current projection up lead i. (b) One example of an SS term in $I(2\Omega)$ similar to (a) with an extra electron scattering event off the potential in lead k. (c) One SV term, similar to (b) except charge pileup at point 1 occurs inside V. (d),(e) The only two processes contributing to $I_i^{VV}(2\Omega)$. The potential acts twice inside V. Note the counterpropagating natures of (d) and (e) ensure their cancellation for $\mathbf{B}=0$.

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